

# Parametric resonance in atomic force microscopy: A new method to study the tip-surface interaction

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## Abstract

We propose a new method to investigate interactions involved in atomic force microscopy (AFM). It is a dynamical method relying on the growth of oscillations via parametric resonance. With this method the second and third derivatives of the tip-surface interaction potential can be measured simultaneously. Because of its threshold behavior parametric resonance AFM leads to sharp contrasts in surface imaging.

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Atomic force microscopy (AFM) is a local method (on the scale from nanometers to microns) able to sense the interaction potential between a relatively flat surface and a more or less well-defined probing tip mounted on a cantilever [1].

The different methods to sense the interaction potential can be divided into two groups: Static methods where the lever deflection proportional to the force between tip and surface is measured [2] and dynamic methods where the frequency shift of the harmonically driven lever is measured [2]. From static methods one can thus get the first derivative of the interaction potential as a function of the tip-surface distance. With dynamic methods where the oscillation amplitude is held small one gets the force gradient, i.e., the second derivative of the interaction potential by measuring the shift of the resonance frequency. There are also dynamical methods with large oscillation amplitudes which involve nonlinear oscillations induced by the interaction [3–5]. In all these dynamical methods the AFM cantilever is excited near of one of its resonance frequencies.

In this Letter we propose a new dynamical method which is able to measure the second and third derivative of the interaction potential simultaneously. Contrary to the dynamical methods described above, the AFM cantilever is excited near a frequency which is roughly *twice* its resonance frequency. First we discuss qualitatively the main properties of such a parametric resonance. In particular, we will see from which kind of measurable data one can obtain the third derivative of the potential. Following this qualitative discussion a single-oscillator model is treated quantitatively. This leads to convenient (but approximate) formulas for the second and third derivative. Finally, we propose an operational scheme for measuring the quantities from which one obtains the derivatives in question.

The reader might ask, can anything interesting happen by driving an AFM cantilever away from its resonance frequency? The answer is: It is possible to rapidly detect the growth of an instability due to the *nonlinear* tip-surface interaction. The instability mechanism is the first-order *parametric resonance* [6]. Parametric resonance occurs in an oscillator which is driven by a *modulation of its eigenfrequency* rather than by an external force. The simplest example is a pendulum with an oscillating length. Such oscillations can be excited very easily

if the modulation frequency is *twice* the average eigen frequency of the oscillator. Everybody knows this intuitively from the experience of a swing-boat at a fair.

How does this mechanism manifest itself in an AFM cantilever which is driven in the usual way? Upon approaching the sample, the resonance frequency of the cantilever changes slightly by an amount proportional to the second derivative of the interaction potential. Thus the resonance frequency of the cantilever depends on the tip-surface distance. If the cantilever is driven out of resonance, it nevertheless oscillates with an amplitude which is of the same order as the driving amplitude [7]. Because of these oscillations the tip-surface distance varies which then leads to a temporal modulation of the resonance frequency. Thus parametric resonance can take place. As an instability mechanism, parametric resonance is a threshold phenomenon. The cantilever starts to oscillate only if the modulation amplitude is larger than some threshold. Moreover, this threshold depends on the ratio of the modulation frequency and the eigen frequency. Its absolute minimum occurs when the modulation frequency is twice the resonance frequency, the so-called first-order parametric resonance condition. The minimal threshold is proportional to the damping constant (i.e., the inverse quality factor) [8].

For a fixed modulation amplitude above the minimal threshold and a varying modulation frequency, one finds a frequency regime where parametric resonance occurs. As shown below, the center of the interval is determined by the second derivative of the interaction potential whereas the third derivative determines the width of the interval. If the modulation amplitude is much higher than the minimal threshold, the effects of damping can be neglected. Then the maximum growth rate (which occurs roughly in the middle of the instability interval) is proportional to the third derivative of the potential. Because it does not depend on the  $Q$  factor, the instability may grow much faster than one would expect from the slow relaxation in a system with a high  $Q$  factor.

After this qualitative discussion we calculate parametric resonance in an AFM by a model which describes the AFM by a single oscillator (see also Fig. 1):

$$\frac{\kappa}{\omega_0^2}\ddot{x} + \frac{\kappa}{\omega_0 Q}\dot{x} + \kappa x - V'(d + h \cos \omega t - x) = 0, \quad (1)$$

where  $\kappa$  is the spring constant,  $\omega_0 \equiv 2\pi f_0$  is the resonance frequency,  $x$  is the tip position relative to its equilibrium value,  $d$  is the tip-surface distance in equilibrium without oscillations,  $h$  and  $\omega \equiv 2\pi f$  are the modulation amplitude and frequency, respectively, and  $V(r)$  is the tip-surface interaction potential. Derivatives of  $V$  are denoted by quotation marks, e.g.,  $V'(r) \equiv dV(r)/dr$ .

The tip-surface interaction potential has in general an attractive and a repulsive part. In principle the proposed parametric resonance method can work in non-contact mode as well as in contact mode. Here we will restrict ourself to the non-contact mode (i.e., the tip is in the attractive part of the potential). In the intermittent or trapping mode analytical approach becomes difficult because tip and sample deformations must be taken into account. Furthermore, the damping is more complicated because it increases significantly upon contact [3].

In our model we use for the attractive part of the tip-surface interaction the following effective potential [9] describing the sum of Van der Waals interactions between a plane sample and a tip with a spherical apex:

$$V(r) = -\frac{AR}{6r}, \quad (2)$$

where  $A$  is the so-called Hamaker constant,  $R$  is the tip radius, and  $r \ll R$  is the tip-surface distance. We have chosen  $A = 2.5 \cdot 10^{-19} \text{J}$  and  $R = 10 \text{nm}$ .

In (1) we have assumed that the tip-surface distance is modulated directly by vibrating the sample vertically. Alternatively one can excite the cantilever at its support which also leads to an oscillation of the tip-surface distance (see Fig. 1). Here we restrict our discussion to the former case because in the single oscillator model both kind of driving are mathematically equivalent. That is, driving the support of the cantilever with the amplitude  $H = h\sqrt{[1 - (\omega/\omega_0)^2]^2 + (Q^{-1}\omega/\omega_0)^2}$  leads to a tip-surface oscillation with amplitude  $h$ .

For  $h \neq 0$  the equation of motion (1) has a solution  $x_h(t)$  with the same periodicity as the modulation term, i.e.,  $x_h(t + 2\pi/\omega) = x_h(t)$  for any  $t$ . Parametric resonance means that

the solution  $x_h$  becomes unstable. That is, a tiny perturbation  $\delta x$  added to  $x_h$  increases exponentially in time. Linearizing (1) in  $\delta x$  and assuming

$$x_h \ll h \ll d \quad (3)$$

we get

$$\frac{\kappa}{\omega_0^2} \ddot{\delta x} + \frac{\kappa}{\omega_0 Q} \dot{\delta x} + (x + V_0'' + V_0''' h \cos \omega t) \delta x = 0, \quad (4)$$

where

$$V_0'' \equiv \left. \frac{d^2 V}{dr^2} \right|_d \quad \text{and} \quad V_0''' \equiv \left. \frac{d^3 V}{dr^3} \right|_d. \quad (5)$$

This linearized equation of motion (4) is the well-known Mathieu equation with a damping term. The Floquet theorem implies that (4) has two linearly independent solutions  $c_1(\omega t)e^{\lambda_1 t}$  and  $c_2(\omega t)e^{\lambda_2 t}$  with  $c_{1/2}(\phi + 2\pi) = c_{1/2}(\phi)$ . The so-called Floquet exponent  $\lambda$  is in general complex. Any solution of (4) is a superposition of these solutions. The solution  $x_h$  is stable if any disturbance  $\delta x$  decays. The stability condition is therefore  $\text{Re} \lambda_{1/2} < 0$ .

In order to calculate the Floquet exponents one has to expand the  $c$  functions into Fourier series. This expansion turns (4) into a set of infinitely many linear algebraic equations for the coefficients of the Fourier series. In the case of weak damping and weak driving, i.e.,

$$Q \gg 1, \quad \text{and} \quad \frac{|V_0'''| h}{\kappa} \ll 1 + \frac{V_0''}{\kappa}, \quad (6)$$

one gets good approximations for the Floquet exponents by restricting the Fourier series to the two leading components. That is, we solve (4) approximatively with the ansatz

$$\delta x = (a_+ e^{i\omega t/2} + a_- e^{-i\omega t/2}) e^{\lambda t} + \text{c.c} \quad (7)$$

Plugging the ansatz into (4) and neglecting terms with  $\exp(\pm 3i\omega t/2)$  leads to two linear homogeneous equations for  $a_{\pm}$  which have a nontrivial solution only if  $\lambda$  is a solution of the characteristic polynomial

$$\left(\frac{V_0''''h}{2\kappa}\right)^2 - \left[\left(\frac{\lambda}{\omega_0}\right)^2 + \frac{\lambda}{\omega_0 Q} + 1 + \frac{V_0''}{\kappa} - \left(\frac{\omega}{2\omega_0}\right)^2\right]^2 - \left(\frac{\lambda}{\omega_0^2} + \frac{\omega}{2\omega_0 Q}\right)^2 = 0. \quad (8)$$

This equation can be solved analytically because it is a second order polynomial in  $(\lambda/\omega_0 + 1/2Q)^2$ . Two solutions are always complex with a negative real part. The two other solutions become real if  $h$  is large enough. One of them is always negative whereas the other one becomes positive if

$$h > h_c \equiv \frac{2\kappa}{|V_0''''|} \sqrt{\left[1 + \frac{V_0''}{\kappa} - \left(\frac{\omega}{2\omega_0}\right)^2\right]^2 + \left(\frac{\omega}{2\omega_0 Q}\right)^2}, \quad (9)$$

where  $h_c$  is the threshold for parametric resonance. Typical growth rates given by the solution of (8) with the maximum real part is shown in Fig. 2. The threshold curve  $h_c$  and curves of constant growth rates are shown in Fig 3.

The minimum value of  $h_c$  is given by  $2Q^{-1}\kappa/|V_0''''|$  which occurs at the parametric resonance condition  $\omega = 2\omega_0\sqrt{1 + V_0''/\kappa}$ . Note that the minimum value of  $h_c$  increase like  $d^4$ . Thus for increasing distance  $d$  the minimum value of the modulation amplitude that is necessary to lead to parametric resonance eventually exceeds the distance itself. Beyond that point which scales like  $Q^{1/3}$  parametric resonance is no longer possible.

For  $h \gg 2Q^{-1}\kappa/|V_0''''|$  the damping term in (8) and (9) can be neglected. Using (9) one finds that parametric resonance occurs for

$$\omega_- < \omega < \omega_+, \quad \text{with} \quad \omega_{\pm} = 2\omega_0 \sqrt{1 + \frac{V_0''}{\kappa} \pm \frac{|V_0''''|h}{2\kappa}}. \quad (10)$$

From measuring  $\omega_{\pm}$  one obtains  $V_0''$  and  $V_0''''$ . The growth rate of a disturbance is zero at  $\omega = \omega_{\pm}$  and has its maximum near  $2\omega_0\sqrt{1 + V_0''/\kappa}$ . The maximum growth rate reads

$$\lambda_{\max} = \frac{|V_0''''|h}{4\kappa} \sqrt{1 + \frac{V_0''}{\kappa}}. \quad (11)$$

Note that the handy formulas (10) and (11) hold only if the assumptions (3) and (6) are fulfilled. The parameters of Figs. 2 and 3 clearly fulfill these assumptions.

In order to measure the (positive) growth rate in parametric resonance AFM we propose the following operation procedure. The procedure switches between two modes of operation: The *on* and the *off* mode. In the *on* mode the system is modulated at roughly twice the resonance frequency. In the response of the cantilever we are looking for oscillations at half of the driving frequency (i.e., near the resonance frequency of the cantilever) because such oscillations will be excited if parametric resonance occur. If there is an instability the amplitude  $a$  of these oscillations increases exponentially and eventually exceeds some predefined value  $a_2$ . Then the system switches into the *off* mode where the modulation is switched off. The parametrically excited oscillation decays until the oscillation amplitude decreases below a value  $a_1 < a_2$  which is of the order of the noise level. Then the system switches back into the *on* mode. By measuring the average duration  $t_{\text{on}}$  of the *on* cycles one gets the growth rate  $\lambda$ . Assuming that  $x$  has to be increased from the noise level  $\sqrt{\langle x^2 \rangle}$  up to  $a_2$  we get

$$\lambda \approx \frac{\ln \left( a_2 / \sqrt{\langle x^2 \rangle} \right)}{t_{\text{on}}}. \quad (12)$$

Because the quality factor  $Q$  in vacuum is very high (i.e., of the order of  $10^4$ ) the decay of the oscillation would take much more time than the excitation. In order to shorten  $t_{\text{off}}$  one can use an external control circuit which brings the cantilever much faster back to equilibrium.

We have simulated this operational scheme by integrating the equation of motion (1) numerically. In order to incorporate the effect of noise we have added on the right hand side of (1) an additive force

$$\kappa \sqrt{\frac{2\langle x^2 \rangle}{\omega_0 Q}} \nu(t), \quad (13)$$

with white noise, i.e.,

$$\langle \nu(t) \rangle = 0, \quad \langle \nu(t) \nu(t + \tau) \rangle = \delta(\tau). \quad (14)$$

To extract the amplitude of the cantilever at half of the driving frequency we used a simple band filter, i.e.,

$$\ddot{y} + 2\gamma\omega_0\dot{y} + \left(\frac{\omega}{2}\right)^2 y = x. \quad (15)$$

For the relative bandwidth we have chosen  $\gamma = 0.05$ . From the solutions of (15) we define an oscillation amplitude

$$a \equiv \gamma\omega_0\sqrt{(\omega y)^2 + (2\dot{y})^2}. \quad (16)$$

For  $x(t) = a_0 \cos(\omega t/2)$  one gets exactly  $a = a_0$ . In our simulation we have modeled the external control in the *off* mode by adding on the right-hand side of (1) the force  $-\kappa\dot{x}/(\omega_0 Q_0)$ . Figure 4 shows an example of such a simulation.

In Fig. 2 the growth rates obtained from such simulations are compared with the analytical result. each square (with error bars) are the average of 30 to 50 *on* cycles. There is a good agreement between the analytical result and the simulation concerning  $\omega_{\pm}$ . The growth rate from the simulation is in general too small because Eq. (12) assumes that the parametrically excited mode has initially the amplitude  $\sqrt{\langle x^2 \rangle}$  neglecting the fact that in the noise there are also contributions from the stable modes. Thus the initial amplitude of the parametrically excited modes is on average less than  $\sqrt{\langle x^2 \rangle}$ .

With our simulations we have shown that parametric resonance AFM is possible. Instability intervals and growth rates can be measured by using our proposed on-off scheme. From this data one gets with the help of (10) and (11) the second and the third derivative of the tip-surface interaction potential. This method should work very well in the non-contact mode. Using this method in imaging we expect sharp contrasts because parametric resonance is a threshold phenomena.

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# FIGURES

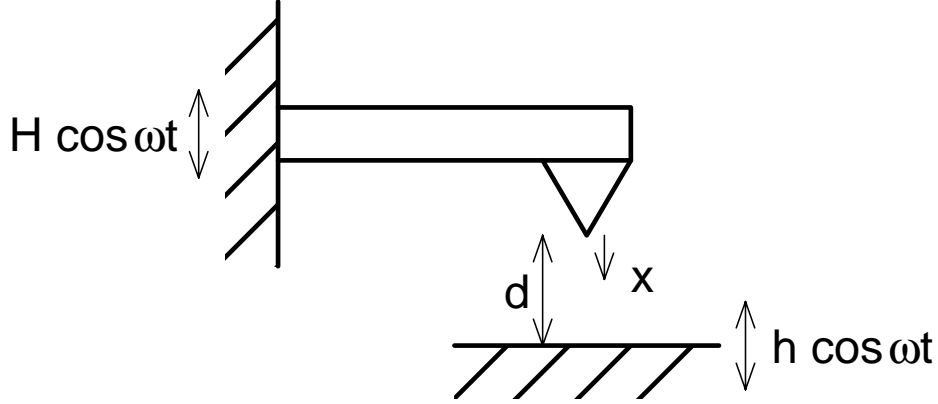


FIG. 1. A schematic sketch of an atomic force microscope (AFM). The tip-surface distance is either modulated directly (modulation amplitude  $h$ ) or indirectly by exciting an oscillation in the cantilever (driving amplitude  $H$ ).

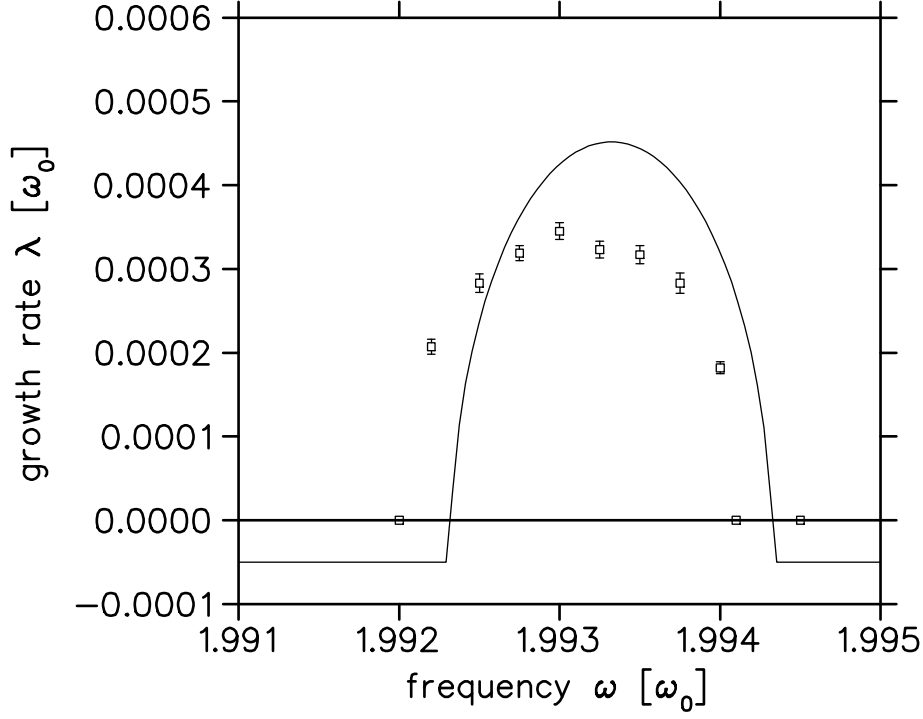


FIG. 2. Growth rate  $\lambda$  as a function of the modulation frequency  $\omega$ . Frequencies and growth rates are measured in units of the resonance frequency  $\omega_0$  of the cantilever far away from the surface. The solid line is the solution of the characteristic polynomial (8). Squares show the result obtained from simulations as shown in Fig. 4. The parameters are  $d = 5\text{nm}$ ,  $\kappa = 1\text{N/m}$ ,  $Q = 10^4$ , and  $h = 0.5\text{nm}$ .

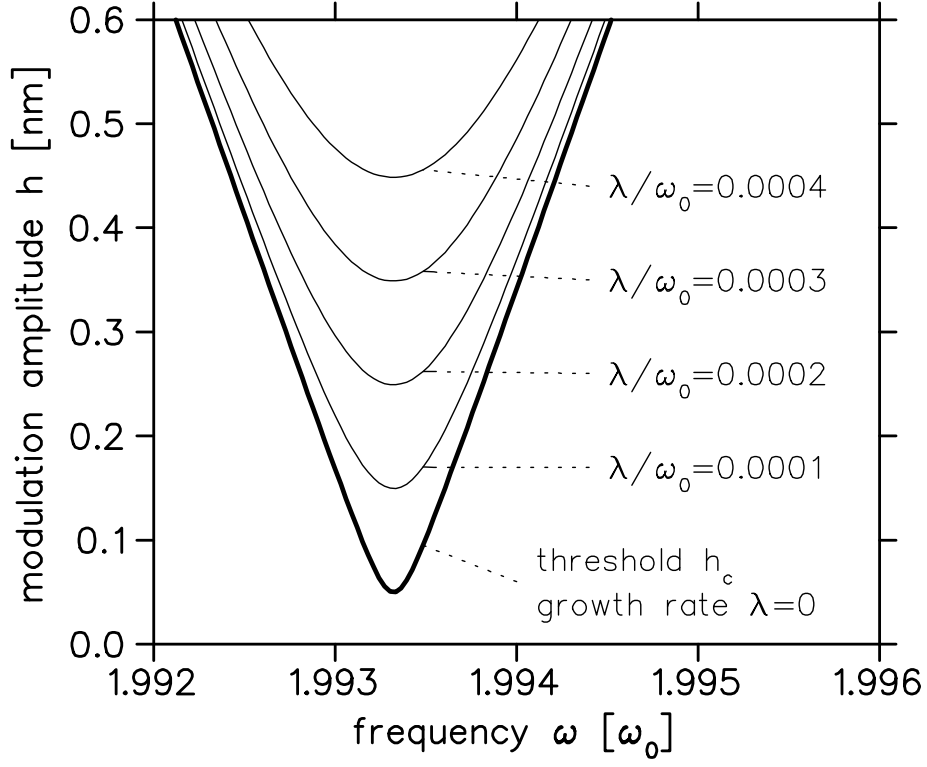


FIG. 3. Instability threshold  $h_c$  and curves of constant growth rate  $\lambda$  as functions of the modulation frequency  $\omega$ . The parameters are the same as in Fig. 2.

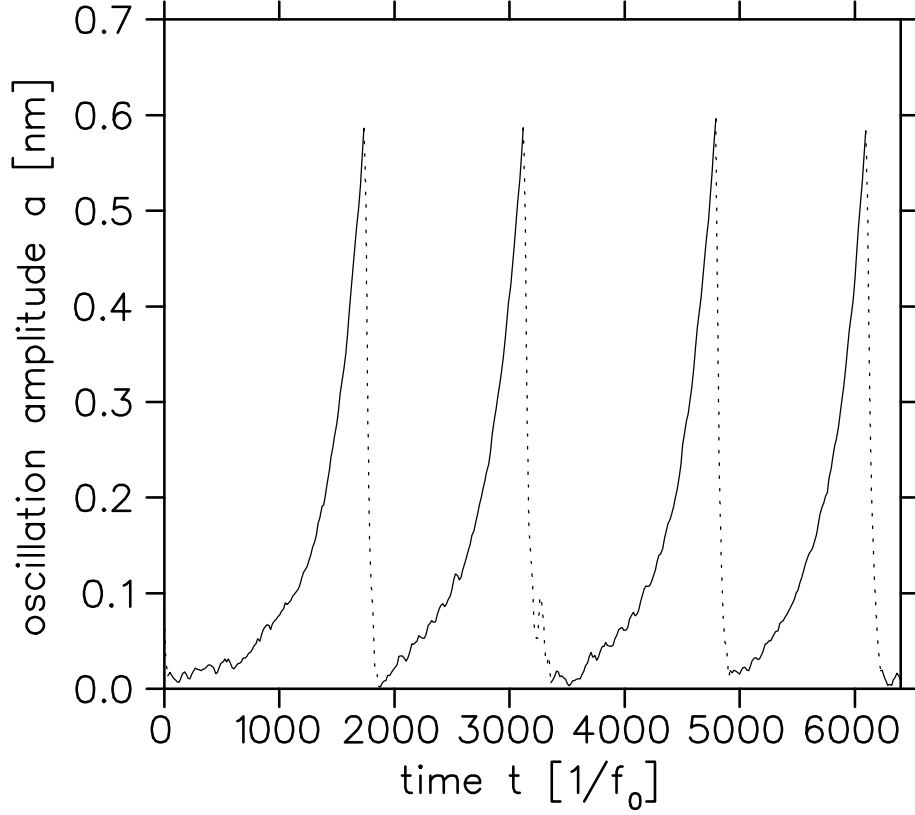


FIG. 4. Simulation of an AFM. Solid lines show the *on* mode where the tip-surface distance is modulated with the frequency  $\omega = 1.993\omega_0$ . This modulation is switched off when the amplitude  $a$  [defined by (16)] reaches a threshold  $a_2$  (here:  $a_2 = 0.6$ ). In the *off* mode (dotted lines) the quality factor  $Q$  is set to a relatively low value  $Q_0$  (here:  $Q_0 = 100$ ). This simulates in a simplified way a control which brings the oscillating AFM back to equilibrium on a fast time scale. When the amplitude  $a$  is below a critical value  $a_1 < a_2$  (here:  $a_1 = 0.01$ ) the system switches back to the *on* mode. The parameters are the same as in Fig. 2. Noise level:  $\langle x^2 \rangle = 10^{-3}$ .